FAST NEUTRON INDUCED FLUX PINNING IN Tl-BASED HIGH-T_c SINGLE CRYSTALS AND THIN FILMS, HIGHLY TEXTURED TAPES AND MELT-TEXTURED BULK 123-SUPERCONDUCTORS

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Abstract

Various compounds (Tl-2223, Tl-1223, Tl-2212) as well as material forms (single crystals, thin films, ceramics, tapes) of Tl-based high temperature superconductors were investigated by magnetic and transport techniques. Tl-2223 has a very "low lying" irreversibility line (H||c) and negligible critical current densities J_c at 77 K. However, the irreversibility line shifts to higher fields and temperatures and J_c is strongly enhanced, even at 77 K, after fast neutron irradiation. In contrast, the related Tl-1223 compound has a much steeper irreversibility line (H||c) similar to that of Y-123. J_c is significant up to 77 K, even in the unirradiated state, and can be largely improved by neutron irradiation. Transport measurements made on Tl-1223 tapes still show much lower critical current densities. Tl-2212 and Tl2223 thin films have J_c's at 77 K, which are comparable to those of Tl-1223 single crystals. Transport measurements on highly textured Bi-2223 tapes as well as flux profile measurements on Nd-123 bulk superconductors confirm the beneficial effects of neutron induced defects (collision cascades) for flux pinning.

1. Introduction

In contrast to classical technical superconductors such as Nb_3Sn, most of the high temperature superconductors show a very wide range in their (H,T)-phase diagram, which is charaterized by a reversible state. Although this opens up the possibility of determining essential mixed state parameters such as the penetration depth of the upper critical field, which is of interest for a fundamental understanding of superconductivity in these materials, many attempts have been made to increase the irreversible state, which is usually very small and separated from the reversible regime by the so called irreversibility line. Only in the irreversible state currents can be carried without dissipation. For applications of superconductors critical current densities J_c<5 10^8 Am^{-2} in fields of at least 2 T are required, but this has not yet been reached in practical materials at liquid nitrogen temperature. Although there is a variety of superconductors with T_c<100 K, they all have one common problem: vortices are only weakly pinned because of the two-dimensional (2D) charcater of these materials and the high temperatures they are used at.

Flux pinning can be improved by subjecting the material to fast neutron irradiation leading to a tremendous increase of J_c and a shift of the irrevesibility line to higher fields and temperatures [e. g. 1]. We will show that this is also the case in Tl-based superconductors. Furthermore, results on one member of the large family of Tl-cuprates, i. e. the one with the 1223 structure with only one Tl-layer in between the Cu-O-layers, will be discussed. Its high irreversibility line, comparable to that found in Y-123 or even higher, and the high J_c-values render this material a candidate for future applications. In this paper, studies of the irreversibility line and Jc on Tl-1223 (ceramic and single crystal) and on Tl-2223 single crystals, in both cases prior to and following neutron irradiation, are presented. They are compared to transport critical current densities measured on Tl-2212 and Tl-2223 thin films and on Tl-1223 tapes. Furthermore, we will briefly discuss enhancements of transport J_c's in highly textured Bi-2223 tapes [2] and of critical current densities in bulk Nd-123 [3] superconductors.

2. Experimental

Single crystals with the nominal composition Tl-1223 were examined by SQUID-magnetometry. For a detailed description of the growth of these compounds, see [4, 5, 6]. The crystals are small plateletes with dimensions $1030 \cdot 430 \cdot 65\ \mu m^3$ (Tl-2223) and $380 \cdot 350 \cdot 108\ \mu m^3$ (Tl-1223), respectively. They were mounted onto small U-shaped aluminium sample holders, which fit into an aluminium rod and allow a reproducible and accurate orientation with respect to the field. Hysteresis loops were measured in an 8T-SQUID magnetometer up to an applied field of 8 T; zero-field-cooled (ZFC) and field cooled (FC) measurements were carried out both in the 8 T- and in a 1 T-SQUID magnetometer with H||c. The superconducting transition temperature was measured in an applied field of 1.1 mT with H||ab. The Tl-2223 single crystal was subjected to fast neutron irradiation (E<0.1 MeV) to fluences of $2 \cdot 10^{21}\ m^{-2}$, $4 \cdot 10^{21}\ m^{-2}$, $8 \cdot 10^{-21}\ m^{-2}$, and $16\ 10^{21}\ m^{-2}$, for the Tl-1223 single crystal only the first three irradiation steps are available at present.

$(TlPb)Sr_2Ca_2Cu_3O_9$-ceramics were prepared as described e. g. in [7]. From the resulting pellet a cubic piece ($2 \cdot 2.5 \cdot 1.84\ mm^3$) was cut and examined by SQUID magnetometry. The average grain size determined from SEM photographs is 4.2 μm.

Measurements of the transport critical current densities were made on Tl-2223 [8] and Tl-2212 [9] thin films as well as on Tl-1223 and Bi-2223 [10] tapes. The epitaxial thin films were deposited onto single crystalline $LaAlO_3$ substrates and patterned to bridge dimensions of 15-28 mm width, 400-6000 mm length, and thicknesses between 150 nm (Tl-2212) and up to 1550 nm (Tl-2223). Large contact pads (~0.5 mm^2) were covered with silver layers. Indium was used as a buffer between the silver contact pins and the film, which resulted in sufficiently low contact resistances. J_c was defined by an electric field criterion of 10 mV/cm. A rotating device allowed us to vary the orientation of the films with respect to the field direction ($\upsilon=0^\circ$: H||ab), but the current was always kept prependicular to the c-axis.

The silver sheated tapes were fabricated by the "powder in tube" (PIT) technique. They were glued onto a small copper plate. This plate is isolated with a thin ceramic layer and has four contact pads for the transport measurements. The sample can be measured in fields up to 6 T at temperatures between 2 K and 150 K. The orientation of the tape in the magnetic field can be varied by rotating the sample holder, keeping the field and current perpendicular to each other. 0° corresponds again to H||ab and 90° to H||c. The angular resolution is better than 1°. The critical current was determined using a standard four probe DC technique and a criterion of 1 mV/cm.

Finally, the bulk 123-superconductors were investigated by the flux profile technique [11] and by ac susceptibility in an 18 T magnet system. The sample were cut into cubes of about $3 \cdot 3 \cdot 3\ mm^3$ with their edges being parallel to the main crystallographic directions.

All the irradiations were made in the central irradiation facility of the TRIGA Mark-II reactor in Vienna, whose flux density distribution has been established accurately [12]. At full reactor power, the flux density of fast neutrons amounts to $7.6 \cdot 10^{16}$ $m^{-2}s^{-1}$ (E>0.1 MeV). The sample temperature during irradiation is not well known, but estimated to be < 50 °C. During irradiation, the samples were kept under He atmosphere in a sealed quartz cylinder, which was inserted into an open aluminium container.

3. Transition Temperature

T_c was measured on all single crystals in the unirradiated state as well as after each irradiation step. Tl-2223 has the highest T_c (121.5 K), which decreases continuously with irradiation (120.5 K, 118 K, 117.3 K 113.5 K). T_c as a function of fluence shows a small plateau at the lowest fluence [13], but generally a linear decrease with a slope of -4.8 K per 10^{22} neutrons/m^2. This decrease is more pronounced than in Y-123 [14]. Tc should reach 0 K at a fluence of 3 10^{23} neutrons/m^2, which is almost three times higher than for Y-123, because of its higher T_c (15). The Tl-1223 single crystal has a much lower T_c of 107 K, which is even lower than values found by other groups for this material [6]. Only the bulk sample and the Tl-1223 tape show transition temperatures similar to published data (114 K and 117 K, respectively). The decrease of Tc with neutron fluence in the Tl-1223 single crystal is almost linear (107 K (unirradiated), 105.2 K (2 1021 m^{-2}), 104.7 K (4 1021 m^{-2}), and 101.7 K (8 10^{21} m^{-2})) with a slope of -6.37 K per 1022 neutrons/m^2, which represents a much stronger decrease than in Tl-2223. Furthermore, the small plateau at the lowest fluences is not found. T_c of the films was found from resistivity measurements (I = 1 μA) to be 111 K (Tl-2223), about 10 K below that of the single crystal, and 107.5 K for the Tl-2212 film. The transition temperatures of the Bi-2223 tape and of the Nd-123 sample are in agreement with published results.

4. Critical current densities J_c of the Tl-superconductors

The critical current densities J_c of the single crystals were calculated as a function of the local induction B using an anisotropic Bean model, which takes demagnetization effects into account [16]. A simple bean model was employed for the determination of the intragrain critical current densities as a function of the applied field for the polycrystalline bulk sample. The grains were assumed to be infinite cylinders. A direct comparasion between J_c of the single crystals and the bulk sample is possible, since the difference between B and moH is very small. Figure 1 shows J_c for the Tl-2223 and Tl-1223 single crystals at 10 K for H||c. J_c of Tl-2223 is strongly temperature and field dependent and disappears above 40 K. Tl-1223 shows fishtails, which are already pronounced at 10 K and keep J_c nearly constant (about $2.3 \cdot 10^{10}$ Am^{-2}) over a wide local induction range at 10 K, at least up to 8 T. Compared to Tl-2223, the temperature dependence of Jc is not so strong, it drops below 109 Am^{-2} only above 60 K and is still significant (about $2 \cdot 10^8$ Am^{-2}) at 0.5 T and 77 K. The polycrystalline Tl-1223 sample does not show a fishtail effect. J_c is higher than in the single crystals and higher than in Y-123 (~10^{10} Am^{-2}), namely 10^{11} Am^{-2} at 5K and 5-$12 \cdot 10^9$ Am^{-2} at 40 K. Even at 93 K hysteresis loops were found, which correspond to a critical current density of $2.7 \cdot 10^8$ Am^{-2} at 0.5 T.

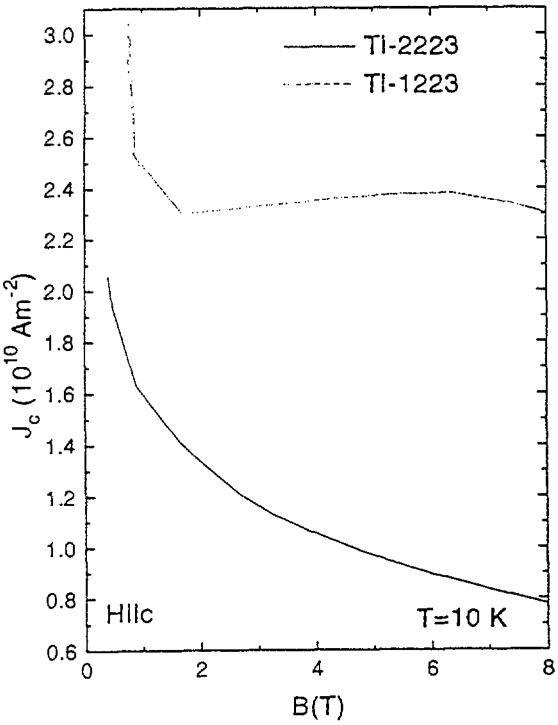

Fig. 1. Comparasion of J_c at 10 K in Tl-2223 and Tl-1223 single crystals.

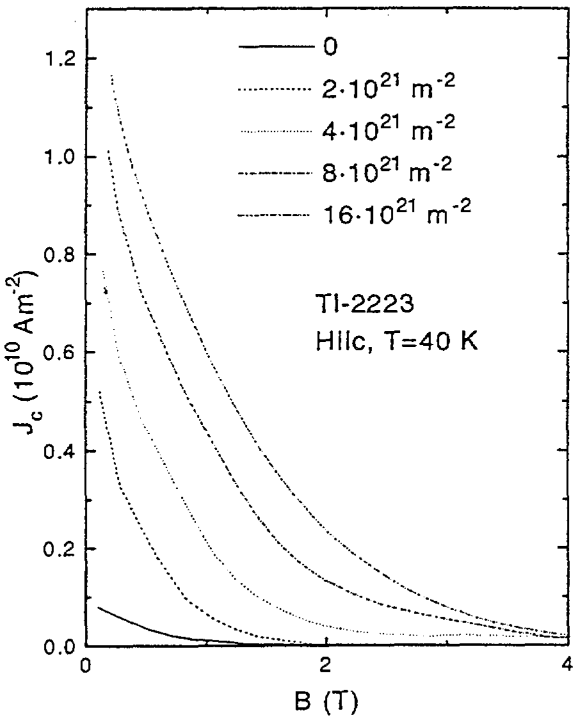

Fig. 2. J_c in a Tl-2223 single crystal irradiated to different fluences.

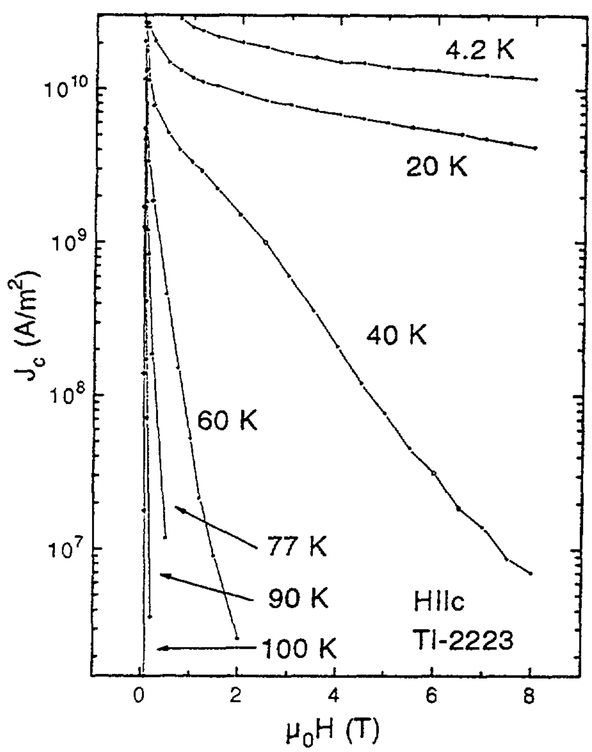

Fig. 3. J_c of a Tl-2223 film.

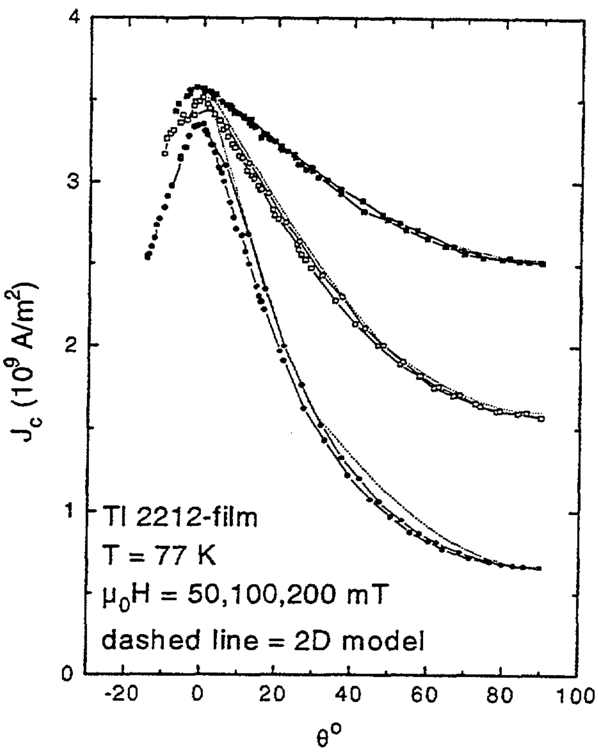

Fig. 4. J_c anisotropy in Tl-2212.

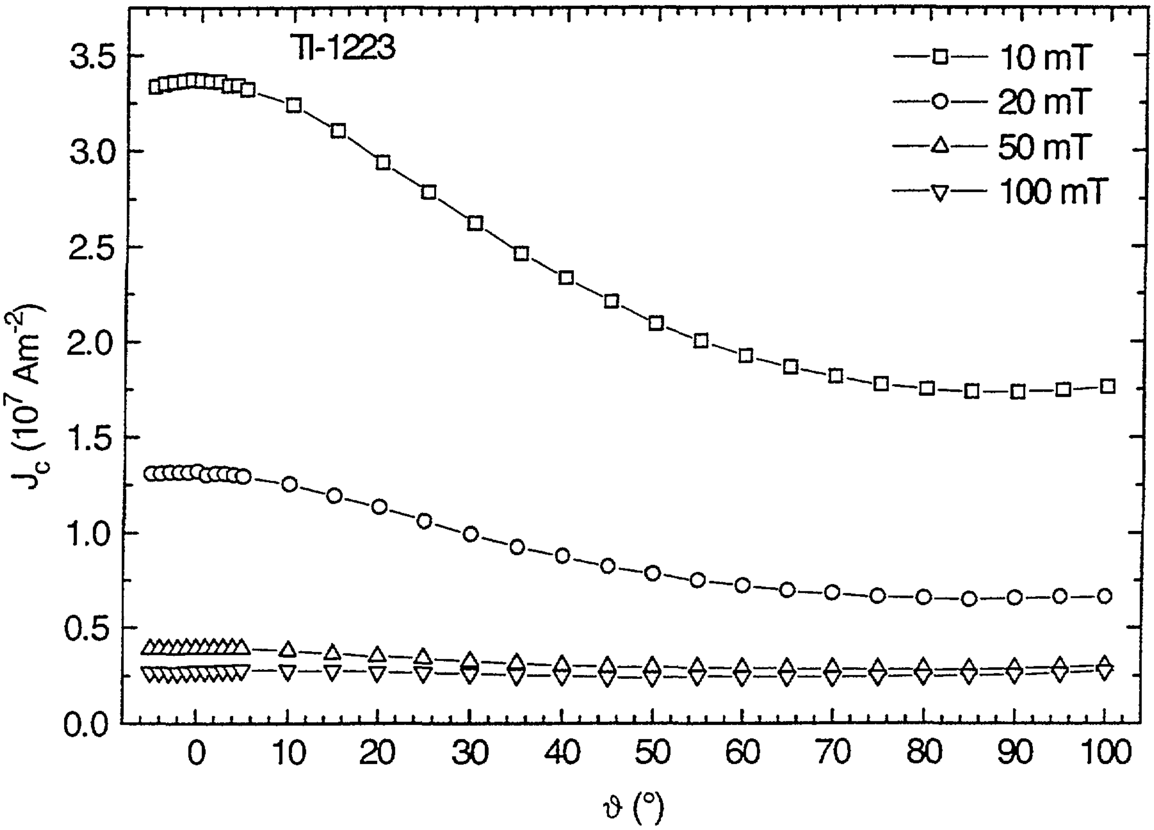

Fig. 5. J_c anisotropy in a Tl-1223 tape at 77 K.

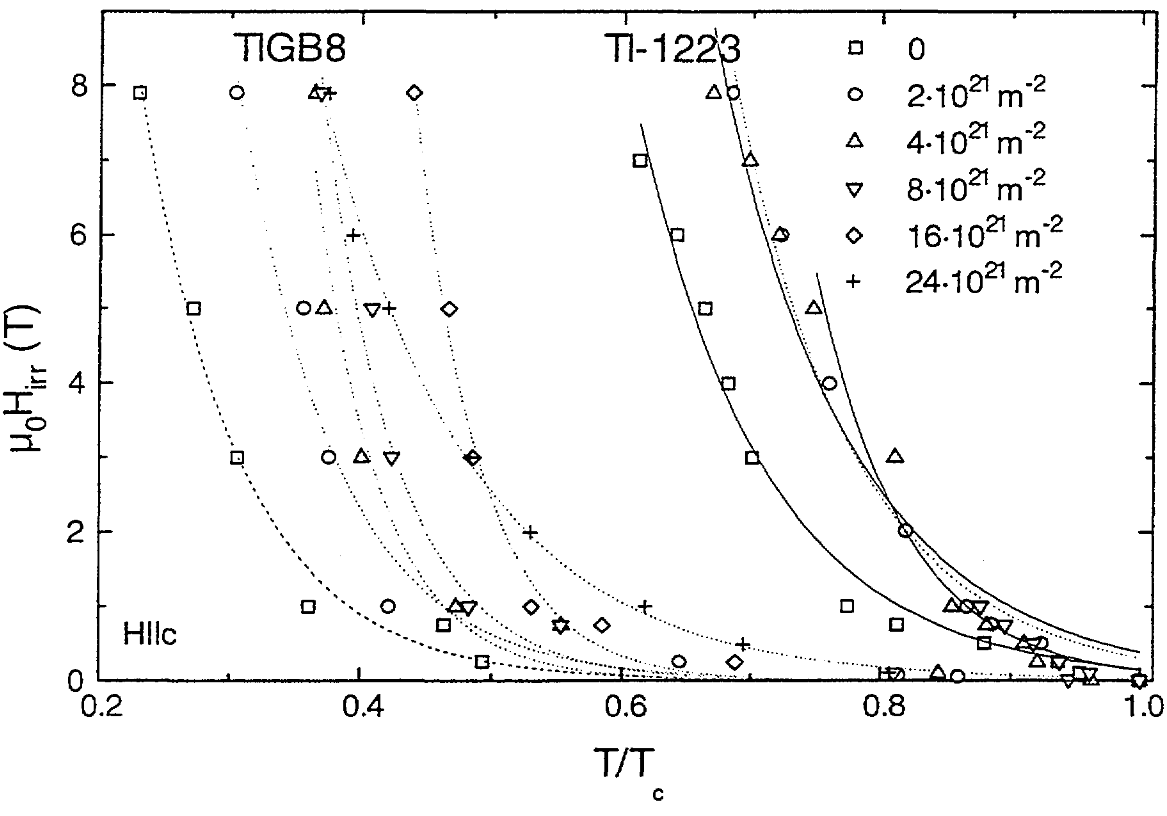

Fig. 6. Irreversibility lines of Tl-2223 and Tl-1223 single crystals.

The best Tl-2223 single crystal was subjected to fast neutron irradiation to cumulative fluences up to 16 10^{21} m^{-2}. The J_c-values increase systematically after each irradiation step. At 1 T and 5 K, J_c changed from about $2.7 \cdot 10^{10}$ Am^{-2} to $5\text{-}6 \cdot 10^{10}$ Am^{-2} after the last irradiation step. At 40 K and 1 T the increase is much more pronounced, from $1 \cdot 10^8$ Am^{-2} to $6.1 \cdot 10^9$ Am^{-2} corresponding to an enhancement factor of 61 (Figure 2). At 77 K, critical current densities above $5 \cdot 10^7$ Am^{-2} are observed for B>0.3 T. After the last irradiation step J_c is of the same order of magnitude of small inductions as for Tl-1223 in the unirradiated state.

Neutron irradiation of the Tl-1223 single crystal led to similar results. The fishtails disappeared after the first irradiation step, leading to a strong enhancement at low and high inductions, respectively. J_c at 77 K and at 0.5 T increased from $2.19 \cdot 10^8$ Am^{-2} (unirradiated) to $1.4 \cdot 10^9$ Am^{-2} ($2 \cdot 10^{21}$ m^{-2}), $1.9 \cdot 10^9$ m^{-2} ($4 \cdot 10^{21}$ m^{-2}), and finally to $2.14 \cdot 10^9$ Am^{-2} after the last irradiation step, i.e. by one order of magnitude. A similar behaviour is found at lower temperatures. At 40 K and 1T J_c increased from $3.23 \cdot 10^9$ Am^{-2} (unirradiated) to $2.5 \cdot 10^{10}$ Am^{-2} ($8 \cdot 10^{21}$ m^{-2}) corresponding to an enhancement factor of 7.7. Even at 93 K J_c-values up to $3 \cdot 10^8$ Am^{-2} are found below 0.2 T after the last irradiation step. At this temperature J_c was zero before irradiation.

Transport critical current densities as a function of the applied field at several temperatures are shown in Figure 3 for a Tl-2223 film. The J_c-values are of the same order of magnitude as in the Tl-2223 single crystal, i.e. $\sim 8 \cdot 10^9$ Am^{-2} (Tl-2223) and $\sim 5 \cdot 10^9$ Am^{-2} (Tl-2212) at 4 T and 20 K. Rotation measurements performed on the Tl-2212 and Tl-2223 films, reveal 2D behaviour [17] (Figure 4), even up to transition temperature. The anisotropy is higher in the Tl-2223 films than in Tl-2212, and the critical current density is much more field dependent.

For the Tl-1223 tape the field dependence of the transort critical current density with the magnetic field applied parallel and prependicular to the tape surface, was measured at 77K and 4.2 K up to 3T. The zero field value of the crytical current density J_c is $1.9 \cdot 10^8$ A/m^2 at 4.2 K and it is 8.4 10^7 A/m^2 at 77 K. However, J_c still drops very rapidly in magnetic fields, and disappears at about 0.2 T at both temperatures. Rotation measurements in fixed magnetic fields were made at the same temperature (Figure 5). The flat behaviour near 0° shows that the sample is poorly textured. Applying a 2D-model to the rotation measurements, the misalignment angle of the grains inside the tape can be obtained [18]. It is found to be approximately 16°, i.e. more than twice as high as in Bi-2223 tapes (cf. Below).

5. Irreveribility lines

For the single crystals and the bulk sample the irreversibility lines were determined using a method based upon the distortion of the SQUID response curve. For a detailed description of this technique see [19, 20]. Results for $\mu_o H_{irr}$ as a function of the reduced temperature $t=T/T_c$ are plotted in Figure 6. The reversible regime of Tl-2223 is very large and extends almost down to $t \approx 0.5$, leading to a very small initial slope of the irreversibility line. A rapid increase occurs below $t \approx 0.3$. Although Tl-1223 is more three dimensional, like for example Y-123, the shape of the irreversibility line is more related to that of the Tl-2223 compound. The form of the irreversibility line can be described by an exponential law $\mu_o H_{irr}(t)=b \cdot \exp(-at)$ with fit parameters a and b.

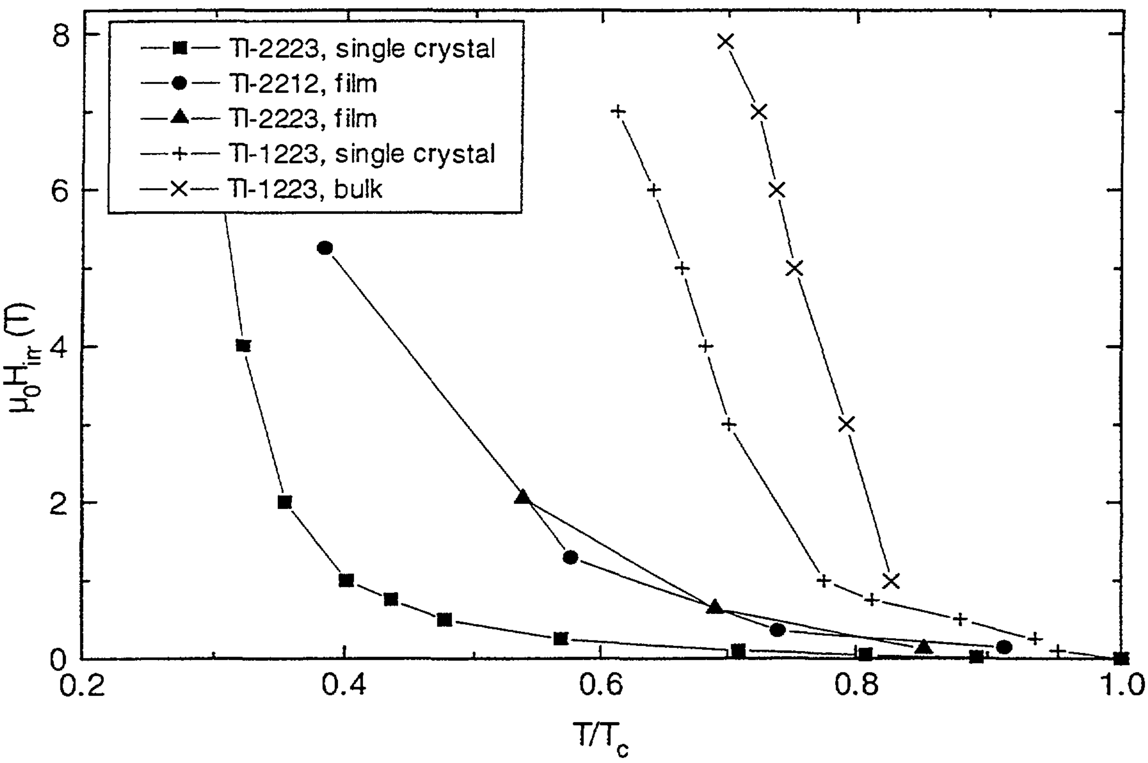

Fig. 7. Comparison of irreversibility lines in single crystals and thin films.

After each irradiation step the irreversibility line of the Tl-2223 single crystal (H||c) shifts to higher fields and temperatures. Between the last two irradiation steps there is almost no change indicating that $16\cdot10^{21}$ m^{-2} is probably the highest fluence the material can sustain without major degradation of superconductivity. Compared to the irreversibility line of Tl-1223 in the unirradiated state, they are still much lower and folow again an exponential law.

Neutron irradiation of the Tl-1223 single crystal has led to the following results (H||c). After the first irradiation step ($2\cdot10^{21}$ m^{-2}) the irreversibility line is shifted to higher fields and temperatures. After the second and third step ($8\cdot10^{21}$ m^{-2}) no further changes are observed within experimental uncertainity. Thus, the increase of the irreversible regime is much more moderate than in Tl-2223. However, since J_c is still enhanced and the decrease of T_c is not dramatic after the third irradiation step, a further moderate shift of the irreversibility line may be expected after subjecting the sample to a fluence of $16\cdot10^{21}$ m^2.

Using a criterion of $4\cdot10^6$ A/m^2, the irreversibility points at high temperatures were also calculated from the I-V characteristics of the Tl-based films. This criterion is somewhat larger than that used for SQUID measurements ($\sim10^6$ Am^{-2}). The irreversibility lines for the Tl-2212 and Tl-2223 films show the same shape as found for the single crystals. However, the rapid increase occurs already at about t=0.5, which is slightly higher than for the Tl-2223 single crystals, but still far below Tl-1223 (see Figure 7).

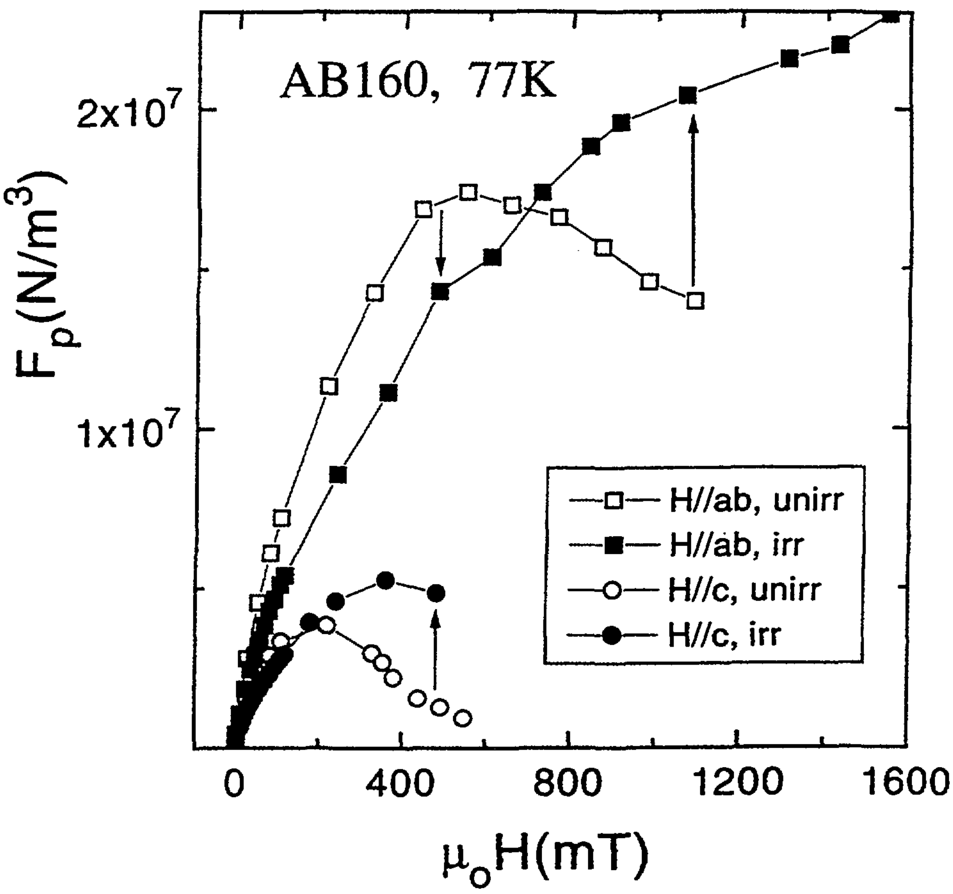

Fig. 8. Volume pinning forces vs applied field prior to and following fast neutron irradiation.

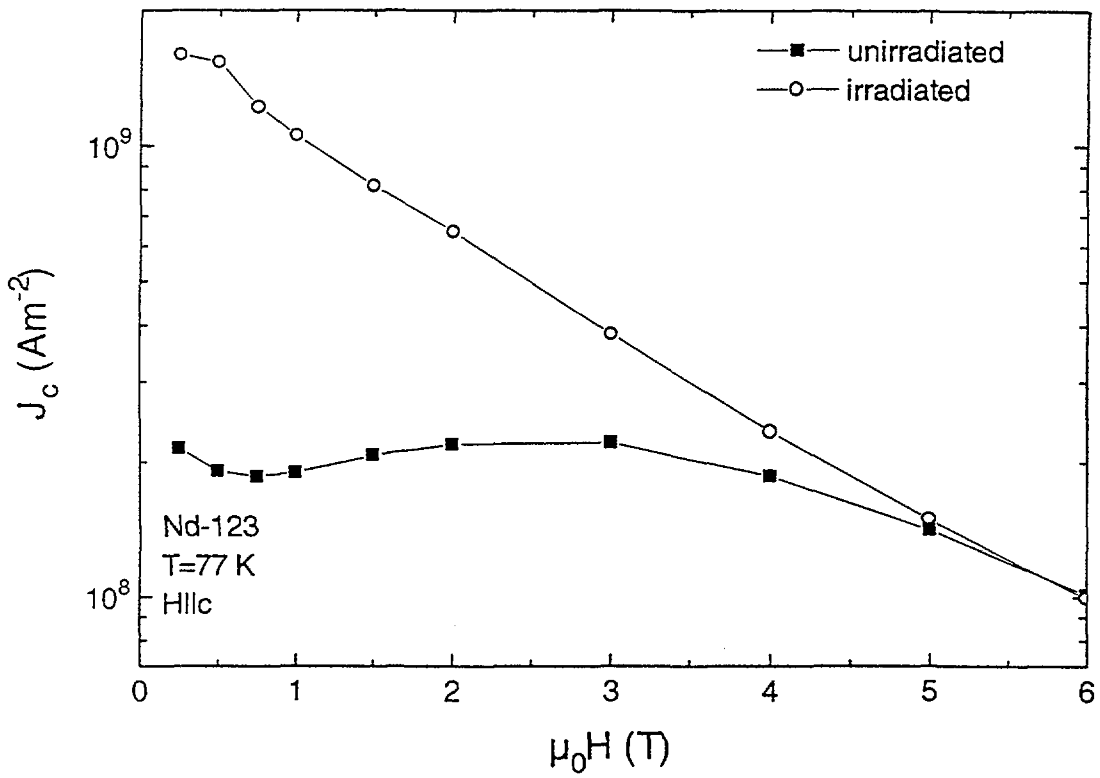

Fig. 9. J_c at 77 K in Nd-123 before and after irradiation.

6. Radiation Effects on Bi-2223 tapes

Since all of the measurements discussed so far, are based on the magnetic response of the sample and further evaluation in terms of some models, a direct transport measurements of J_c following irradiation seemed to be of considerable interest. The corresponding experiment was done on a well textured Bi-2223 tape prepared by the powder in tube technique. After characterization in the unirradiated state, the tape was irradiated to a fluence of $4 \cdot 10^{21}$ m^{-2} (E>0.1 MeV). Because of the high level of radioactivity induced in the silver sheath, a waiting time of four months was needed prior to the experiment. Typical results for the volume pinning forces at 77 K are shown in Figure 8. We note that both for H||c and for H||a,b, the critical current densities decrease at low magnetic fields, but then significantly increase beyond the levels in the unirradiated state. The initial degradation in the low field regime is ascribed to radiation-induced damage of the weak link structure, which has been generally observed in transport experiments on irradiated ceramics. However, once the weak link dominated field regime is surpassed, the experimental data clearly show, that flux pinning of pancake and/or Josephson vortices is obviously improved by the defects introduced by fast neutron irradiation. This holds for both major orientations of the tape surface with respect to the magnetic field. Furthermore, from a complete measurement of the angular dependence of J_c we find, that J_c still scales with the c-component of the magnetic field, i. e. that the superconductor is still in a optimization studies of this material, because it demonstrates the necessity of not only improving the grain alignment within the tapes, but also of enhancing the flux pinning capability within individual grains.

7. Critical Current Densities of Nd-123 Bulk Superconductors

Considerable progress has recently been achieved by synthesizing Nd-123 bulk materials under reduced oxygen atmosphere [3]. This leads to an enhancement of T_c by approximately 2 K compared to the standard Y-123 phase. Furthermore, a strong "fishtail" feature is introduced into the material (Figure 9), which keeps the critical current density at a relatively high level up to magnetic fields of about 5 T, in marked contrast to melt textured Y-123, where this field is close to the irreversibility point at 77 K. In fact, the irreversibility field (H||c) reaches 9 T in Nd-123 at the boiling point of liquid nitrogen. Neutron irradiation of such a compound to a relatively low fluence ($2 \cdot 10^{21}$ m^{-2}) has led to rather spectacular results as shown in Figure 9. Because of the radiation-induced defects, the fishtail behavior is completely removed and the critical current density pushed up by a factor of ~10 to approximately $2 \cdot 10^9$ Am^{-2} for $H \rightarrow 0$. In present considerations for applications of these bulk materials, such data for J_c and $\mu_0 H_{irr}$ indeed push the frontiers for their application potential quite far ahead. Further work on the subject is currently under way.

SUMMARY

Various compounds as well as material forms of Tl-based high temperature superconductors were investigated. The critical current densities of Tl-2223 can be strongly enhanced and the irreversibility lines shifted to high temperatures and fields to fast neutron irradiation. However, the J_c values found in the related Tl-1223 compound and the position of its irreversibility line cannot be reached. Neutron irradiation of Tl-12223 single crystals has led to an irreversibility line, which exceeds that of Y-123 at 77 K. J_c in the presently available Tl-tapes is still very small and strongly field dependent, thus indicating weak link limitations for the current transport. Tl-2212 and Tl-2223 films show critical current densities which are of the same order of magnitude as those for single crystals 2D behaviour is found up to the transition temperature. Attempts to improve their flux pinning capability by neutron and heavy ion irradiation are currently under way. With regard to textured materials, such as Bi-2223 tapes or Nd-123 bulk superconductors, the successful defect modification by neutron-induced collision cascades demonstrates a high potential for material optimization programs by suitable metallurgical defects.

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References

[1] H. P. Wiesinger, F. M. Sauerzopf, H. W. Weber, H. Gerstenberg, G. W. Crabtree, Europhys. Lett. **20** (1992) 541.

[2] Q. Y. Hu, H. W. Weber, F. M. Sauerzopf, G. W. Schulz, R. M. Schalk, H. W. Neumuller, S. X. Dou, Appl. Phys. Lett. **65** (1994) 3008.

[3]S. I. Yoo, N. Sakai, H. Takaichi, T. Higuchi, M. Murakami, Appl. Phys. Lett. **65** (1994) 633.

[4] G. Brandstätter, F. M: Sauerzopf, H. W. Weber, W. Mexner, H. C. Freyhardt, A. Aghaei, F. Ladenberger, E. Schwarzmann, in "Applied Superconductivity", ed. By H. C. Freyhardt, DGM Informationsges. Verlag, 1993, 109.

[5] K. Winzer, Annalen der Physik **1** (1994) 479.

[6] K. Aihara, T. Doi, A. Soeta, S. Takeuchi, T. Yuasa, M. S. Eido, T. Kamo, S. Matsuda, Cryogenics **32** (1992) 936.

[7] W. Mexner, S. Heede, K. Heinemann, H. C. Freyhardt, B. Ullmann, F. Ladenberger, E. Schwarzmann, in "Critical Currents in Superconductors", ed. By H. W. Weber, World Scientific, Singapore, 1994, pp. 513-516.

[8] S. Huber, M. Manzel, H. Bruchlos, S. Hensen, and G. Muller, Physica C **244** (1995) 337.

[9] S. L. Yan, L. Fang, Q. X. Song, J. Yan, Y. P. Zhu, J. H. Chen, S. B. Zhang, Appl. Phys. Lett. **63** (1993) 1845.

[10] Q. Y. Hu, H. W. Weber, S. X. Dou, H. K. Liu, H. W. Neumuller, J. Alloys Comp. **195** (1993) 515.

[11] R. W. Rollins, H. Kupfer, W. Gey, J. Appl. Phys. **45** (1974) 5392.

[12] H. W. Weber, H. Bock, E. Unfried, L. R. Greenwood, J. Nuc. Mat. **137** (1986) 236.

[13] G. Brandstätter, F. M. Sauerzopf, H. W. Weber, A. Aghaei, E. Schwarzmann, Physica C **235-240** (1994) 2797.

[14] F. M. Sauerzopf, H. P. Wiesinger, H. W. Weber, G. W. Crabtree, Phys. Rev. B **51** (1995) 6002.

[15] M. C. Frischherz, M. A. Kirk, J. Farmer, L. R. Greenwood, H. W. Weber, Physica C **232** (1994) 309.
[16] H. P. Wiesinger, F. M. Sauerzopf, H. W. Weber, Physica C **203** (1992) 121.
[17] G. Samadi Hosseinali, R. M. Schalk, H. W. Weber, A. Ponninger, S. Proyer, P. Schwab, Physica B **194-196** (1994) 2357.
[18] Q. Y. Hu, R. M. Schalk, H. W. Weber, H. K. Liu, R. K. Wang, C. Czurda, S. X. Dou, J. Appl. Phys. **78** (1995) 1123.
[19] M. Suenaga, D. O. Welch, R. Budhani, Supercond. Sci. Technol. **5** (1992) 3.
[20] F. M. Sauerzopf, H. P. Wiesinger, H. W. Weber, G. W. Grabtree, Advances in Cryog. Engineering **38** (1992) 901.